PHYSICAL REVIEW LETTERS

Volume 34

28 APRIL 1975

NUMBER 17

¹D Autoionization Series in Helium*

D. Burch

Department of Physics, University of Washington, Seattle, Washington 98195

and

J. Bolger and C. Fred Moore Center for Nuclear Studies, University of Texas, Austin, Texas 78712 (Received 6 March 1975)

Ten states of doubly excited He have been observed in a high-resolution electron-emission spectrum from 30-MeV O^{5+} +He collisions. Five lines can be identified as the $(2pnp)^{1}D$ + series. The excitation mechanism is discussed and compared with results of an identical measurement using 4-MeV proton excitation.

During the past ten years there has been a great deal of research devoted to the study of doubly excited states in He-both electrons excited to bound states in neutral He. The excitation energies lie above the first ionization energy of He, and thus the states decay primarily by autoionization to the ground state of He⁺ with the emission of a monoenergetic electron characteristic of the excited state. Intensive theoretical work on these states has occurred because of the opportunity to study the strong influence of electron-electron correlation in a simple system,¹ an effect which prohibits the classification of autoionization states by a single configuration.² Further interest has arisen as a result of the relevance of these states to other areas of physics: resonant electron scattering, photoabsorption, impurity resonances in solids, ion-atom scattering, and recombination in astrophysical and laboratory plasmas (see reviews by Burke,³ Smith,⁴ and Massey, Burhop, and Gilbody⁵).

The first insight into the nature of these states came from their systematic experimental observation in electron-energy-loss⁶ and photoabsorption spectra.⁷ The extensive work of Madden and Codling⁷ established several Rydberg series² converging on the n = 2, 3, and 4 levels of He⁺. Although photoabsorption work is limited to ¹P excitation, other series (¹S and ³P) converging on n = 2 were soon observed by Rudd⁸ in the electronemission spectrum of 75-keV H₂⁺ + He collisions. Members of these series were subsequently observed by several groups in electron excitation.⁹ Excitation of autoionization states in keV He⁺ + He collisions has also been recently studied by Gerber, Morgenstern, and Niehaus.¹⁰ Radiative transitions from doubly excited states are observed in beam-foil excitation.¹¹

The first member of the ¹D series is well established experimentally, but excitation methods used previously are not efficient in exciting the remainder of the series. Theoretical predictions of the ¹D energies have been given by Cooper *et al.*¹² In this Letter we report a high-resolution autoionization spectrum of He produced by 30-MeV O⁵⁺ ions as an example of a new means of studying these states. In particular, we present the energies of the $(2p np)^{1}D$ +series (n = 2 to 6), which is strongly excited in this collision, and we discuss the unique features of this means of excitation.

The electron spectra presented here were measured with an apparatus described earlier.¹³ The incident O^{5+} beam of 0.8 μA and 3 mm diameter crossed a diffuse atomic beam of He at a distance of 4 mm from a 0.3-mm-diam gas nozzle. Target pressures were roughly estimated to be a few millitorr; the ambient pressure several centimeters from the jet was 2×10^{-4} Torr. The spectrometer entrance slits were located 1 cm from the scattering center with an acceptance angle of 0.93° (effective solid angle of 2.6 msr viewing a 3-mm path length). The pressure inside the spectrometer chamber was $< 1 \times 10^{-6}$ Torr. Electrons emitted at 90° to the beam direction were energy analyzed with a double-focusing spherical analyzer (McPherson ESCA-36, power supplies: Fluke 3330B Calibrator) and detected with a channel electron multiplier. The electron-energy regions of interest were scanned in 4.57-meV steps at a rate of typically one step per 1.2 μ C of



FIG. 1. Electron-emission spectra at 90° from proton and oxygen-ion collisions with He. Autoionization lines in the oxygen-excited spectrum are identified in Table I. integrated beam current. Repeated scans were stored and added to improve the statistics. The resolving power of the measurements was limited to 1.6×10^{-3} (~60 meV) by collision-related effects—estimated to be dominated by recoil kinematic broadening. Since the observed electron energies depended upon the target gas pressure (see also Refs. 8–10), the spectra were each accumulated at a constant pressure found to be stable to within a few percent by measuring the number of electrons per microcouloumb of beam current without incrementing the analyzer voltages.

Electron spectra are shown in Fig. 1; the O^{5+} excited spectrum is compared to an identical measurement using 4-MeV proton excitation. The spectra shown required ~7 h of data aquisition each. The excitation energies indicated are the observed electron energies plus the ionization potential of He (24.587 eV). An expanded picture of the region near the series limit is given in Fig. 2. The data are summarized in Table I; energies listed are the peak centroid positions normalized to the theoretical value of Bhatia¹⁴ for the $(2s2p)^{1}P$ transition (60.149 eV). The uncertainties indicated are derived from Gaussian fits to the peak shapes and do not include possible systematic errors. The ^{1}D identifications are based upon the calculations of Cooper et al.¹² Although the last three lines of the ${}^{1}D$ series are nearly degenerate with members of the ^{1}P series,



FIG. 2. Electron-emission spectrum at 90° from 30-MeV O^{5+} collisions with He in the region near the series limit of 65.4 eV.

TABLE I.	Autoionization	states	in helium	produced
by 30-MeV c	xygen ions.			

Relative intensity	Experimental energies (eV)	¹ D ^a	¹P b	¹ S ^c
97(14)	57.850(16)			57.842
1000(20)	59.948(03)	60.025		
470(17)	60.149()		60.149	
110(9)	62.130(04)			62.134
233(12)	63.575(04)	63.575		
78(10)	63.690(06)		63.677	
17(8)	64.157(28)			64.182
122(10)	64.472(05)	64.429		
49(8)	64.851(09)	64.797		
33(8)	65.049(14)	64.990	·	

^aCooper et al., Ref. 12.

^bBhatia, Ref. 14.

^cBurke and Taylor, Ref. 15.

it is assumed that the relative ${}^{1}D$ to ${}^{1}P$ intensities are the same as observed for the first two transitions and therefore the peak positions are dominated by the ${}^{1}D$ contributions.

The interference phenomenon apparent in the proton-excited spectrum has been studied in detail recently by several groups for electron, photon, and lower-energy proton excitation.¹⁶

Further measurements were also made with incident oxygen beams of 30-MeV O⁸⁺ and 15-MeV O⁴⁺. These spectra appear nearly identical to the 30-MeV O⁵⁺ spectrum with the only exception that the peak-to-continuum electron-count ratio is slightly improved in the O⁸⁺ spectrum and somewhat worse in the O⁴⁺ spectrum. This may be accounted for by electron loss from the projectile.¹⁷

There are three distinct features of the He autoionization spectrum produced by oxygen ions which reflect upon the excitation mechanism. (1) Only singlet states are produced (see Table I); (2) of these the ${}^{1}D$ series is by far the most prominent; and (3) in contrast to proton excitation, no interference with the continuum is observed. The first point is consistent with the O⁸⁺ data which demonstrate that the excitation mechanism does not involve a one-step electron exchange and, moreover, is not influenced by the projectile electrons in any manner.

The second point, strong *D*-state excitation, suggests—in contrast to excitation by fast protons (analogous to single-photon absorption)—that the strong interaction between the He atom and the oxygen nucleus may proceed by a two-step process. The Coulomb-excitation selection rules would then apply to each electron transition separately, favoring a double E1 excitation to nplevels. At comparable collision velocities, the two-step process would be enhanced by Z^4 (~4000) in the oxygen-ion collisions. We should add, however, that at low proton energies (<100 keV) the ¹D excitation has been observed to be comparable to the ¹P.¹⁸ In the oxygen collisions, the relative intensity ratio $(2p^1)^1D/(2p^2)^1S$ is 4.2 ± 0.4 after correction for the transition energy factor (65.4 $-E)^{3/2}$. This value is near 5=2J+1, as would be expected for independent electron excitation.

The third point, lack of continuum interference, could be accounted for if the nonresonant continuum states were dominated by the *double* ionization of He. The autoionization states would then not overlap with the continuum states which give rise to the observed continuous electron distribution and interference would not be observed. This interpretation is suggested by related collision studies using high-energy heavy ions where multiple ionization is consistently found to be much more probable than single ionization.¹⁹ Again in contrast, multiple ionization by energetic protons is very rare and interference can be expected in these collisions. This interpretation requires further verification, e.g., interference phenomena are strongly dependent upon emission angle; but independent of the origin of the effect, the lack of interference considerably simplifies the extraction of the resonance energies especially near the series limit.

In summary, the autoionization states of He have instigated a large body of fruitful theoretical research¹⁵; the present work shows that still further aspects of the excitation and decay of these states are experimentally accessible through the relatively new technique of atomic collisions with very energetic heavy ions.

We thank D. Holtkamp, K. Roberts, and J. Whitenton for assistance with the experiment and Professor M. E. Rudd, Professor J. Macek, and Professor P. G. Burke for valuable comments.

^{*}Work supported in part by the U. S. Atomic Energy Commission, the U. S. Air Force Office of Scientific Research, and the Robert A. Welch Foundation.

¹U. Fano and C. D. Lin, in *Proceedings of the Eighth International Conference on the Physics of Electronic and Atomic Collisions. Invited Lectures and Progress Reports*, edited by B. C. Cobić and M. V. Kurepa (In-

stitute of Physics, Belgrade, Yugoslavia, 1973), p. 229. ²J. W. Cooper, U. Fano, and F. Prats, Phys. Rev. Lett. 10, 518 (1963).

³P. G. Burke, Advan. Phys. 14, 521 (1965).

⁴K. Smith, Rep. Progr. Phys. 29, 373 (1966).

⁵H. S. W. Massey, E. H. S. Burhop, and H. D. Gil-

body, *Electronic and Ionic Impact Phenomena* (Oxford) Univ. Press, London, England 1969), Vol. 1, Chap. 9. ⁶S. M. Silverman and E. N. Lassettre, J. Chem.

Phys. <u>40</u>, 1265 (1964); J. A. Simpson, G. E. Chamberlain, and S. R. Mielczarek, Phys. Rev. <u>139</u>, A1039 (1965).

⁷R. P. Madden and K. Codling, Phys. Rev. Lett. <u>10</u>, 516 (1963), and Astrophys. J. 141, 364 (1965).

⁸M. E. Rudd, Phys. Rev. Lett. <u>13</u>, 503 (1964), and <u>15</u>, 580 (1965); M. E. Rudd and D. V. Lang, in Proceedings of the Fourth International Conference on the Physics of Electronic and Atomic Collisions. Abstracts (Science Bookcrafters, Inc., Hastings-on-Hudson, N. Y., 1965), p. 153.

⁹N. Oda, F. Nishimura, and S. Tahira, Phys. Rev. Lett. <u>24</u>, 42 (1970); H. Suzuki, A. Konishi, M. Yamamoto, and K. Wakiya, J. Phys. Soc. Jpn. <u>28</u>, 534 (1970); K. Siegbahn *et al.*, *ESCA Applied to Free Molecules* (North-Holland, Amsterdam, 1969), pp. 26 and 149; W. Mehlhorn, Phys. Lett. <u>21</u>, 155 (1966).

¹⁰G. Gerber, R. Morgenstern, and A. Niehaus, J. Phys. B: Proc. Phys. Soc., London <u>6</u>, 493 (1973).

¹¹H. G. Berry, J. Desesquelles, and M. Dufay, Nucl. Instrum. Methods <u>110</u>, 43 (1973); H. G. Berry, to be published. ¹²J. W. Cooper, S. Ormonde, C. H. Humphrey, and

P. G. Burke, Proc. Phys. Soc., London <u>91</u>, 285 (1967). ¹³D. L. Matthews, B. M. Johnson, J. J. Mackey, L. E. Smith, W. Hodge, and C. F. Moore, Phys. Rev. A <u>10</u>, 1177 (1974).

¹⁴A. K. Bhatia, Phys. Rev. A 10, 729 (1974).

¹⁵In addition to the above references see P. G. Burke and A. J. Taylor, Proc. Phys. Soc., London <u>88</u>, 549 (1966); J. Macek, J. Phys. B: Proc. Phys. Soc., London <u>1</u>, 831 (1968); L. Lipsky and A. Russek, Phys. Rev. <u>142</u>, 59 (1966); P. G. Burke and D. D. McVicar, Proc. Phys. Soc., London <u>86</u>, 989 (1965); D. R. Herrick and O. Sinanoğlu, Phys. Rev. A <u>11</u>, 97 (1975).

¹⁶F. D. Schowengerdt and M. E. Rudd, Phys. Rev. Lett. <u>28</u>, 127 (1972); N. Stolterfoht, Phys. Lett. <u>37A</u>, 117 (1971); N. Stolterfoht, D. Ridder, and P. Ziem, Phys. Lett. <u>42A</u>, 240 (1972); A. Bordenave-Montesquieu, A. Gleizes, M. Rodiere, and P. Benoit-Cattin, J. Phys. B: Proc. Phys. Soc., London <u>6</u>, 1997 (1973); V. V. Balashov, S. S. Lipovetskii, and V. S. Senashenko, Zh. Eksp. Teor. Fiz. <u>63</u>, 1622 (1972) [Sov. Phys. JETP 36, 858 (1973)].

¹⁷D. Burch, H. Wieman, and W. B. Ingalls, Phys. Rev. Lett. 30, 823 (1973).

¹⁸M. E. Rudd, private communication of unpublished work.

¹⁹B. Burch, in *Proceedings of the Eighth International Conference on the Physics of Electronic and Atomic Collisions. Invited Lectures and Progress Reports*, edited by B. C. Cobić and M. V. Kurepa (Institute of Physics, Belgrade, Yugoslavia, 1973), p. 97.

Sub-Doppler Spectroscopy of Xenon Laser Lines with a Saturated-Amplification Method. Resolution up to the Natural Width

Ph. Cahuzac and R. Vetter

Laboratoire Aimé Cotton, Centre National de la Recherche Scientifique, 91405-Orsay, France (Received 30 January 1975)

A saturated-amplification method has been applied to the spectroscopic study of several infrared laser lines of xenon. The experiment is characterized by the use of two lasers. Linewidth at very low pressure, isotope shift, and hyperfine structure have been measured.

We report here ultrahigh-resolution measurements of atomic structures by means of a twolaser saturated-amplification method. Saturatedabsorption techniques were first extensively used in molecular spectroscopy for which homogeneous widths are very weak^{1,2}; in atomic spectroscopy, although lifetimes of excited levels are generally short and induce larger homogeneous widths, the gain in effective resolution remains important and a few experiments have been performed.^{1,3,4} We have studied several laser lines of xenon in the near infrared. For these lines, we have observed saturated absorption at high pressures but also saturated amplification at low pressures; both situations are fundamentally equivalent in their principle. In order to obtain the best possible resolution, we have systematically performed the experiments at pressures as low as possible, i.e., in saturated amplification; then the homogeneous width is nearly equal to the natural one and is very weak compared to